Ultrafast solvation dynamics explored by nonlinear optical spectroscopy

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Chapter 2

Cavity-dumped femtosecond Titanium:sapphire laser: 
*The light source for non-linear optical spectroscopy*

Part of the work as presented in this chapter is covered by the following papers:

Maxim S. Pshenichnikov, Wim P. de Boeij, and Douwe A. Wiersma,


Maxim S. Pshenichnikov, Wim P. de Boeij, and Douwe A. Wiersma,

Wim P. de Boeij, Maxim S. Pshenichnikov, Koos Duppen and Douwe A. Wiersma,
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Abstract

A femtosecond cw-mode-locked Ti:sapphire laser is constructed that generates pulses as short as 10.5 fs, with an average output power of 500 mW. Direct application of the laser toward non-linear spectroscopic investigations is hampered by its high repetition rate, high average power and relative low energy per pulse. It is demonstrated that the cw-mode-locked laser can be cavity-dumped, allowing the generation of 13-fs, 60-nJ pulses, at repetition rates up to 200 kHz. The laser can also withstand higher dumping repetition rates, although with lower dumping efficiencies. Intracavity spectral filtering allows generation of tunable (from 750 to 860 nm) pulses, with durations around 25 fs. The advantages of the cavity-dumped laser over the cw-mode-locked laser are demonstrated in a series of non-linear optical experiments. The intensity of the cavity-dumped pulses is more than sufficient to induce third-order (and higher order) non-linear optical effects in dissolved dye molecules. On injection of the output of the cavity-dumped laser into a single-mode polarization-preserving fibre, a white-light continuum is generated. This continuum is recompressed to a pulse with duration of 7.5 fs. The cavity-dumped Ti:sapphire laser seems to be an excellent near-IR light source for ultrafast non-linear optical experiments.
2.1 Introduction

In physics, chemistry and biology a vast range of processes can be distinguished, that occur on nano-, picosecond and even femtosecond time scales.\textsuperscript{[1-3]} Given the importance of such processes, it is not surprising that a better understanding of the dynamical features has motivated the development of suitable laser sources for probing these transient events.\textsuperscript{[1-3]}

The ultimate time-resolution for time-domain spectroscopic experiments can never be better than the inverse bandwidth of the excitation light used, as given by the fundamental restriction of the Heisenberg uncertainty relation. A light source providing the experimentalist with a potentially short optical pulse, must therefore inherently accommodate a large spread in photon frequencies.

For the generation of ultrashort laser pulses, all the frequency modes should have a well-defined phase relation with respect to each other. In this sense, the concept and implementation of mode-locking techniques in laser sources has been crucial.\textsuperscript{[4,5]} Developments in laser technology, making use of solid state gain materials such as Nd-YAG and Nd-glass, have lead to the generation of pulses in the picosecond range.\textsuperscript{[6]} Later on, the large bandwidth gain media supplied by the dye laser materials, in combination with different mode-locking techniques have pushed the pulse durations down to the sub-picosecond domain.\textsuperscript{[7]}

In 1981, with the invention of the colliding-pulse mode-locked (CPM) dye laser by Fork \textit{et al.}, it was demonstrated that pulses with durations of sub-100 fs could be generated.\textsuperscript{[8]} Further shortening of the pulses was inhibited by the residual group delay velocity dispersion (GVD) of the pulse inside the laser cavity. Inclusion of an active control over the group velocity dispersion, by means of a 4-prisms negative dispersion element in the cavity, resulted in the generation of 27-fs pulses from a colliding-pulse mode-locked laser.\textsuperscript{[9,10]} Amplification of the CPM laser pulses in combination with fibre-chirping and prism-grating compression techniques led to the generation of laser pulses as short as 6 fs.\textsuperscript{[11]} The prism-controlled CPM laser was the workhorse in many laser laboratories for ultrafast studies in all branches of sciences during the 80’s.\textsuperscript{[2]}

A new era in the solid state laser field was born, with the characterization of a laser material based on the titanium doped sapphire crystal, Ti:Al\textsubscript{2}O\textsubscript{3}.\textsuperscript{[12]} Shortly after the characterization of the optical parameters, the successful cw-lasing action in this material was demonstrated.\textsuperscript{[10]} The material characteristics of Ti:sapphire, namely a broad gain bandwidth, a large energy storage density, and excellent thermal properties, make it an excellent candidate to be used in femtosecond laser and amplification systems.

The potential of short pulse generation was first demonstrated employing the additive pulse mode-locking mechanism.\textsuperscript{[14-23]} In this case the main cavity is coupled to an external resonator, containing an optical fibre. After the radiation has experienced a non-linear phase
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modulation in the fibre, the laser beam is coupled back to the main cavity. This mode-locking mechanism allows the generation of sub-picosecond pulses. The use of the coupled cavity inherently necessitates an interferometric stable feedback mechanism, thereby lowering the stability and robustness of this laser design. Other mode-locking techniques, such as passive mode-locking (saturable absorber dye),\textsuperscript{22,23} regenerative mode-locking,\textsuperscript{24,25} and passive mode-locking by means of semiconductor saturable absorbers in the cavity,\textsuperscript{26} were also employed for the successful generation of sub-100-fs pulses.

A major breakthrough in the femtosecond laser field was established by Sibbett et al. in 1991, with the discovery of the action of self-mode-locking.\textsuperscript{27} Also referred to as Kerr-lens mode-locking (KLM), the method relies on the non-linear refractive index properties ($n_2$) of the Ti:sapphire crystal. The self-focussing action in the crystal in combination with an intra-cavity aperture results in intensity dependent losses and causes the mode-locking of the laser. Implementation of the KLM mode-locking, and accurate control over the group velocity dispersion allow the generation of short femtosecond pulses, as has been demonstrated in many laboratories.\textsuperscript{27-38} Pulse durations in prism-controlled Ti:sapphire oscillators are limited to 10 fs, as a result of the inherent presence of uncompensated higher order dispersion terms.

With the realization of the Kerr-lens mode-locked lasers, the design of an oscillator that generates femtosecond optical pulses has reached a stage of remarkable simplicity and robustness. The ability of the Ti:sapphire solid-state oscillator to generate 10-fs pulses\textsuperscript{37,38} as well as its excellent reliability, make it an attractive ultrafast light source in the laboratory. Not surprisingly, the class of lasers based on Ti:sapphire has rapidly gained ground in the field of ultrafast spectroscopy.\textsuperscript{2}

New developments in the methods to compensate for the group velocity dispersion inside the laser cavity, thereby employing chirped dispersion mirrors, have lead to the generation of ultrashort pulses with sub-10-fs pulse durations.\textsuperscript{19} Similar results have been obtained for Ti:sapphire lasers including a semiconductor saturable absorber system.\textsuperscript{40} Although the gain bandwidth the Ti:sapphire crystal can support the generation of pulses shorter than 4 fs, technical limitations in the bandwidth of laser mirrors\textsuperscript{41} and spectral non-uniformities in the resonator beam profile\textsuperscript{42} currently limit further shortening of the pulses.

Apart from the tremendous progress in the Ti:sapphire laser oscillator design, great strides have also been made in the amplification of pulses from the oscillator.\textsuperscript{43-50} be it that the output pulse currently is limited to about 18 fs as a result of spectral distortions and dispersive effects in the amplifier.\textsuperscript{45,51} Multi-pass low repetition rate amplifiers (1 Hz up to 20 Hz) generating pulses at the (milli)Joule level,\textsuperscript{43-45} regenerative amplifiers with kHz pulse rates,\textsuperscript{46-49} and high repetition rate (sub-MHz) amplifier systems\textsuperscript{50} are nowadays routinely used in many laboratories.

Although the state-of-the-art amplifier can provide the experimentalist with extremely powerful pulses at various repetition rates, there is a wide range of (nonlinear) ultrafast optical experiments, for instance photon-echo and pump-probe, that would greatly profit from
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a Ti:sapphire-based light source that would combine the advantages of a very short light pulse, as provided by the oscillator, with medium-high peak power at variable repetition rates. The latter issue is important to avoid effects of accumulation in single-shot type optical experiments. It is well known that the incorporation of a cavity-dumper in the laser resonator is a simple and convenient method to realize these goals. This technique has been successfully used with femtosecond hybridly mode-locked dye lasers. For example, the generation of pulses as short as 48 fs with pulse energies of 20 nJ has been reported using such a system. Ramaswamy et al. were the first to report on a cavity-dumped Ti:sapphire laser that produced 50-fs pulses with energies of 100 nJ at variable repetition rates. They also demonstrated that stable mode-locking of the laser persisted despite the strong perturbations caused by the dumping process.

Following the results of Ramaswamy et al., the question arises whether cavity-dumping can be achieved with a pulse length provided by a state-of-the-art Ti:sapphire oscillator. In this chapter it is demonstrated that cavity-dumping of a self-mode-locked Ti:sapphire laser can be extended to the 10-fs time domain. This is accomplished without sacrificing much either of the ultrashort pulse duration or of any other characteristics of the original Ti:sapphire laser, although the pulse energy increases by an order of magnitude compared to the cw 10-fs laser.

In Section 2.2 the Kerr-lens mode-locking principle, pulse formation mechanism and resonator stability conditions will be addressed. As an intermediate step to the cavity-dumped version of the laser, in Section 2.3 the design of the conventional cw-mode-locked Ti:sapphire laser is presented. The cavity-dumping of the mode-locked Ti:sapphire laser is addressed in the Section 2.4. Characteristics of the laser operation and the dumped laser pulses will be given. In Section 2.5 it is shown that with a minor modification, the cavity-dumped laser can deliver tunable laser pulses. Finally, the potential of the cavity-dumped Ti:sapphire laser toward non-linear optical experiments will be demonstrated in Section 2.6.

2.2 Kerr-lens mode-locking, pulse formation, and resonator stability

In the following section three important aspects of the operation principles of the Ti:sapphire laser will be addressed. The Kerr-lens mode-locking (KLM) is discussed that causes the pulsed operation of the laser, and also leads to suppression of the possible cw-wave. Since the femtosecond solid-state Ti:sapphire laser can be classified as a solitary laser, the formalism of pulse formation in this class of lasers is presented. Finally the stability conditions of the employed resonator will be treated.
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2.2.1 Kerr-lens mode-locking

The Kerr-lens mode-locking mechanism (KLM) is directly related to the self-focussing action of the Ti:sapphire crystal.\textsuperscript{[20]} The effect of the intensity dependent lens on the intracavity beam is schematically depicted in Figure 2.1. Upon an increase in the intensity of the input beam, the beam size of the output beam will decreases accordingly. An intracavity aperture transfers this intensity dependent beam size modulation into a subsequent modulation in the propagation losses. For a properly aligned cavity, a smaller attenuation is achieved for the high intensity circulating pulse mode. The competing low intensity cw-mode experiences a larger attenuation and is usually suppressed under normal operation conditions. In Figure 2.1, the pinhole acts as a filter between both modes of operation. This method is referred to as the hard aperture mode-locking. Besides the hard aperture, a second discrimination mechanism is always present, and results from the fact that the pump beam exciting the gain medium forms an intrinsic (soft) aperture (spatial gain modulation). Cavity modes that have the largest overlap with the excited gain medium experience the largest gain. The KLM mode-locking constitutes the necessary mechanism for the initial build up of the laser pulse in the cavity. The steady state laser conditions are closely connected to the concept of soliton propagation in non-linear media, and will be discussed in the next section.

2.2.2 Pulse formation in solitary laser systems

The pulse formation mechanism of the Kerr-lens mode-locked lasers relies on the so-called soliton pulse formation mechanism. A full account on this mechanism can be found in numerous papers,\textsuperscript{[35,38,58-60]} here we will present the most important elements of the theory, as they are relevant to this chapter. In the description of the pulse formation and pulse propagation in the laser resonator, two important mechanisms can be distinguished, self-phase modulation (SPM) and group delay dispersion (GDD).

The self-phase modulation (SPM) is based on the intensity dependent refractive index ($n_2$) of the Ti:sapphire crystal. Besides the afore mentioned action of self-focussing, the same

![Fig. 2.1. All-optical modulator based on the Kerr-lens induced in a medium with positive $n_2$. The optical beam travels from left to right. The high power beam (solid line) at the output has a smaller diameter than the low power mode (dashed curve), and is not attenuated by the aperture. (Adapted from Ref. 35).](image-url)
non-linearity affects the phase properties of the electric field. Intensity changes at the leading
and falling edge of the pulse, result in the generation of new frequency components in the
pulse.

The second mechanism is the process of group delay dispersion (GDD). As a result of
the wavelength dependent refractive index of the medium (n=n(λ)), modes with different
frequencies propagate at different velocities. In general, it is this effect that results in the
broadening of a short pulse as it propagates through a medium with non-zero GDD.

The combined and balanced action of the self-phase modulation (SPM) and the group
delay dispersion (GDD) is the basis for the generation of a short pulse. In the soliton
formation mechanism the change of the pulse as it passes through the cavity elements is often
described by an operator-like formalism. The aforementioned mechanisms, GDD and SPM
constitute the different operators. In the description, the cavity mode is represented as an
amplitude modulated plane wave:

\[ E(t) = v(t)e^{i\omega t} \] (2.1)

where \( v(t) \) is the complex time-dependent envelope and \( \omega \) the optical carrier frequency.
Under the condition that the Kerr non-linearity is the dominant mechanism, a stationary
solution for the envelope function \( v(t) \) can be obtained:

\[ v_s(t) = \left( \frac{W}{2\tau_s} \right) \text{sech}\left( \frac{t}{\tau_s} \right)e^{i\varphi} \] (2.2)

where \( W \) denotes the pulse energy, and \( \tau_s \) is the pulse duration of the solitary solution. The
phase \( \varphi \) is the phase shift the pulse accumulates as it makes a full round trip through the
cavity, and depends on the dispersive properties of the resonator:

\[ \varphi = \varphi_0 + n \frac{D}{\tau_s^2} \] (2.3)

The initial phase of the pulse is given by \( \varphi_0 \), \( D \) denotes the net group delay dispersion, and
\( n \) labels the number of round trips. From Eq.(2.2) it can be inferred that the pulse intensity
envelope is described by a quadratic hyperbolic secant (sech^2(t)).
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The above formalism furthermore predicts, in the limit of near-fundamental soliton propagation, the pulse duration $\tau$, in terms of cavity constants to be:

$$\tau = \frac{3.53 D}{\phi W} + \alpha \phi W$$  \hspace{1cm} (2.4)

In Eq.(2.4) $\phi$ denotes the strength of the Kerr non-linearity, causing the action of the SPM. The coefficient $\alpha$ (ranging from 0.1 to 0.25) depends on the position in the cavity. The shortest pulse duration is obtained for a cavity configuration with a minimum net group delay dispersion. The net GDD should be negative in order to remain in the stable pulse propagation regime. Higher order terms in the group delay dispersion (not included in the above analysis) will lead to pulse distortions and eventually tend to break up the pulse.\textsuperscript{[61-62]}

The above treatment complies well with the experimental results as found in pulse generation conditions for pulses longer than 15 fs. Recently the propagation of ultrashort laser pulses, have been modelled.\textsuperscript{[61-65]} The calculations are based on models invoking (3D,t) calculations and take the actual cavity configuration in consideration.\textsuperscript{[66]} The space-time focussing conditions of the circulating pulse as it enters and passes through the Ti:sapphire crystal, imposes some fundamental limitations to the generation of ultrashort pulses. The crucial factor in the generation of ultrashort pulses is formed by the inherent dispersion of the Ti:sapphire medium. By minimizing the length of the Ti:sapphire crystal (at the expense of the total gain), shorter pulses are predicted.\textsuperscript{[65]}

2.2.3 Stability zones of the laser

Besides an active gain medium, the optical resonator forms the second crucial element in the design of the laser. The optical resonator must be constructed in such a way that upon several passes through the resonator, the radiation remains captured, and amplification of the radiation is accomplished. This condition results in certain ranges where the parameters of the laser configuration can be changed. The stability zones for this configuration can be calculated making use of the ABCD matrix formalism. For a full account on this matter the reader is referred to Ref. 67.

The layout of a four-mirror Ti:sapphire cavity is depicted in Figure 2.2a and the calculated stability configurations are depicted in Figure 2.2b. The calculations are based on the parameters, (length dimensions and curvatures of mirrors) as encountered in the actual experimental setup (Section 2.3 and Figure 2.3). The length of both arms extending from the folded section ($L_{23}$, $L_{41}$) is kept constant as well as the radius of curvature of the focusing...
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Fig. 2.2. (a) Layout of the configuration as used in the cavity calculations. The distances between the mirrors are $L_{12} = 101$ cm, and $L_{14} = 60$ cm. M1, M2 - r=10 cm curved mirrors; M3, M4 - flat high reflecting mirrors; $\theta$ - folding angle of the resonator; Ti:S - Ti:sapphire crystal, $L_c=4$ mm. (b) Stability zones of the cw-mode-locked laser, as a function of the distance between the two curved mirrors around the Ti:sapphire crystal, $L_{12}$. The stability zones I and II for cw-operation are indicated by the gray areas. The dashed lines mark the boundaries of the stability zones in case an additional (Kerr) lens is simulated ($f=30$ mm). The cw-laser mode shapes for the operation in the center ($\gamma$) as well as near the limit of stability zone II ($\beta$) are depicted in the Figure. For comparison the modeshape for femtosecond mode-locking operation ($\alpha$) is given as well. For a discussion on these mode-shapes see Section 2.3.2. Mode profiles were measured by a CCD camera, and the intensity is displayed logarithmically to enhance the low intensity parts of the mode.

cavity mirrors. The running variable in the calculations is formed by the distance between the two curved mirrors ($L_{12}$). In Figure 2.2b two regions of stability can be distinguished. For historical reasons, these regions are referred to as stability zone I and II and are separated by a region where the laser resonator is unstable. The appearance of an unstable gap is a result of a laser configuration with unequal length arms ($L_{12}$, $L_{14}$).

In the calculations the non-linear (Kerr) induced phenomena is initially neglected, results are thus predicted for cw-laser operation. For modelling the self-focussing action of the Ti:sapphire crystal, a positive lens is placed inside the Ti:sapphire crystal. With inclusion of the Kerr-lens, significant changes in the location of the stability zones are found. Especially the second stability zone shifts to smaller values $L_{12}$. Although the strength of the Kerr-lens is taken somewhat arbitrarily, from this simple analysis one can already conclude that when
the laser operates near the inner border of the second stability zone a preferential gain should exist for the high-intensity mode-locked mode. Note that this effect is only based on the cavity stability conditions in combination with the effect of self-focussing, no effect of an aperture on the cavity beam is included here.

In a theoretical study, Brabec et al. presented an analysis on the hard aperture Kerr-lens mode-locking and its relation towards the cavity design. In their conclusion, they specify the following design procedure for the optimization of the Kerr-lens mode-locking performance. The folding angles (θ, Figure 2.2) should be chosen such that the astigmatism is compensated. An asymmetry in the cavity configuration should be introduced, where one arm of the laser is made longer than the other. A hard aperture must be placed in the short arm of the cavity, near the end mirror (HR). Two possible regimes of operation can be distinguished that exhibit a relative large differential (intensity dependent) gain factor. The laser resonator can be aligned to operate in either the first (I) or second (II) stability zone, but should in both cases be aligned near the inner border (Fig 2.2). The conclusions from a similar study presented by Georgiev et al. corroborate the predictions of Brabec et al. It furthermore seems that the hard aperture method of mode-locking, necessitates the operation of the laser in the first stability zone.

As discussed before, the soft aperture, as a result of the spatial gain modulation, is inherently present in the laser. In a study presented by Piché and Salin, the stability of the cavity was studied, where the effect of self focussing and gain saturation were included. Under certain conditions, the combination of self-focussing and gain saturation can produce a differential gain that favours the growth of short pulses and eliminates cw oscillations. The soft aperture discrimination mechanism eliminates the requirement to insert a physical hard

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**Fig. 2.3.** Schematic of the cw-mode-locked Ti:sapphire laser. Ar$^+$ - argon-ion laser (Coherent Innova 310); L - f=12.5 cm lens; Ti:S - 4 nm Ti:sapphire crystal (Union Carbide); HR - high reflector (CVI); M1-M2 - R=10 cm mirrors (Spectra-Physics); M3 - pump steering mirror; M4 - Mirror gold-coated; OC1, OC2 - T=10 % output coupler (LEZER-OPTIKA); P1-P4 - fused silica prisms; Output 1 - Mode-locked laser output; Output 2 - cw-laser output; PS - Periscope for beam steering and polarization rotation.
aperture in the laser as a mode filter. For non-symmetrical cavities (one arm is longer than the other) a substantial differential gain was predicted when the laser is operated near the limits of the stability zones.\cite{70}

### 2.3 Cw-mode-locked Ti:sapphire laser

In this section the design of the cw-mode-locked femtosecond Ti:sapphire laser will be discussed.\cite{71-73} The cavity layout as well as specific aspects of some of the optical elements in the cavity will be addressed. Finally the cw-mode-locked operation characteristics of the laser will be presented.

#### 2.3.1 Layout of the laser

The layout of the design is given in Figure 2.3. The laser configuration consists of a 4-mirror resonator with one folded section. The optical resonator encloses two curved dichroic mirrors, transparent for the pump light and highly reflective for the laser radiation. Two flat mirrors close the linear cavity, one acting as a high reflector and one as an output coupler. The two arms extending from the folded section are asymmetric. The longest arm encloses the dispersion compensation prisms. Two possible configurations can be chosen, depending on the position of the prism P1. For mode-locked operation the prism P1 is positioned in the cavity beam (close to the clipping position), thereby deflecting the beam towards the prism P2 and the output coupler OC1. For alignment purposes, the prism P1 is retracted from the beam and the second output coupler (OC2) is used. Both arms (M1-P1-P2-OC1 and M1-OC2) have the same optical length, ensuring identical stability conditions.

A 4 mm long, 0.15\% doped and Brewster cut Ti:sapphire crystal is placed in the folded section. The overall absorbance of the incident laser power (Ar\textsuperscript{+} laser) is about 80\%. Given the low absorption cross-section\cite{12} and (compared to dye laser materials) a relative low gain cross-section,\cite{35} a long gain medium is required to ensure efficient pumping and overall gain. As a result of this length, the crystal must be pumped collinearly to obtain the best overlap between pump and resonator mode. An anti-reflection coated lens is used for focusing the Ar\textsuperscript{+} pump beam in the Ti:sapphire crystal. The polarization of the pump beam is rotated by a periscope. The polarization of the laser mode is parallel to the optical table and governed by the lossless propagation on the Brewster surfaces of the crystal and the prisms, as well as the orientation of the pump laser and the laser crystal. To avoid the accumulation of heat in the Ti:sapphire crystal, the supporting mount is cooled by running tap water. Under normal operation conditions, the temperature of the crystal is slightly higher than room temperature, which furthermore prevents the accumulation of condensation on the crystal.
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As a result of the presence of the Brewster angle Ti:sapphire crystal, some astigmatism is introduced in the cavity. However, for this one can compensate by tilting the folding mirrors M1 and M2 to a certain extend. Based on the expressions presented by Kogelnik et al., the angle $\theta$ (Figure 2.2) can be calculated as:

$$\theta = 2 \arccos \left[ -C/2 \sqrt{(C/2)^2 + 1} \right]$$

$$C = \frac{t (n^2 - 1) \sqrt{(n^2 + 1)}}{n^3 R}$$

(2.5)

where $t$ is the thickness, and $n$ the refractive index of the Brewster angle medium, and $R$ is the radius of curvature of the folding mirrors. With a refractive index of $n=1.76$ and a thickness of $t = 4$ mm, a folding angle of $\theta = 15.2^\circ$ is found. For the output couplers, different values for the transmission coefficient can be used, ranging from $T=1\%$ up to $T=20\%$. In the presented design a $T=10\%$ output coupler is implemented.

The negative group delay dispersion (GDD) element, necessary to sustain the pulse generation is formed by a double-pass prism arrangement. The Brewster angle fused silica prisms are aligned for minimum deviation. The choice of fused silica is based on the restriction that the prism pair can compensate the positive GDD of a 4 mm long Ti:sapphire crystal with a reasonable prism separation distance. Furthermore, in comparison with other types of glasses, fused silica exhibits a low amount of third-order dispersion. To compensate for the Ti:sapphire crystal and the inherent passage through the glass of the prism pair, a prism separation of 54 cm is needed.

The laser beam is coupled out at the dispersive end of the laser and is passed through a double prism arrangement to recollimate the dispersed beam. The presence of the extra-cavity prism pair allows the pre-compensation of dispersive optics in the following autocorrelator or experimental setup.

For pumping the Ti:sapphire laser a cw Ar$^+$ion laser (Coherent Innova 310, Powertrack) is employed. The typical pump power (all Ar$^+$ lines) is in the range from 4.5 to 6.5 Watt, but optimal performance was obtained for a pump power of 5.5 Watt.

2.3.2 Operating characteristics of the cw-mode-locked laser

Without the insertion of the prisms, and making use of the second output coupler (OC2), the laser produces around 1 Watt of cw-radiation centred around 800 nm, when pumped by 5.5 Watts. A conversion efficiency (absorbed pump power over laser power) of more than 20% (output coupler is $T=10\%$) can be easily achieved in the cw-regime. The insertion of the prism pair in the long arm of the resonator and extracting the radiation through OC1, reduces
the cavity by a few percent, as a result of the additional losses by the double passage through the prism pair.

The Ti:sapphire rod may act as a birefringent filter in the cavity and thus works as a mode selector. Only the modes that have the correct polarization orientation will propagate without any losses. Setting of the c-axis of the sapphire crystal parallel to the polarization direction of the circulation radiation, results in the lossless propagation over the whole accessible gain bandwidth.\cite{71,72} Careful alignment of the crystal axis is crucial to sustain the broadband mode-locked operation of the laser.

For both the predicted positions, (inner borders of the stability zones, Sec. 2.2.3 and Figure 2.2b) mode-locking was experimentally verified. The most stable mode-locking was found at the inner boundary of stability zone II. No hard aperture was needed when operated at this position. Calculations of Piché et al. that include the soft aperture gain effects, also predict the largest differential (mode-locked over cw) gain to be expected at this point.\cite{70}

When the laser operates near the inner boundary in the second (II) stability zone, it generates a (cw) output power of 300 mW. As was depicted in Figure 2.2b, the output mode shape of the laser change as the cavity configuration is changed from operation in the centre of stability zone II toward the operation near the inner edge. As is common in this arrangement, the near Gaussian transverse laser mode changes into slightly egg-like shape. Upon the mode-locking of the laser this mode changes back to a circular symmetric shape and a significant increase in the laser power is observed (up to 500 mW) as a result of a preferential gain effect for the short pulse mode as discussed in Section 2.2.3. The mode-locking was initiated by either tapping the mirror at the dispersive side of the cavity (OC1) or by moving the intra-cavity prism P2 back and forth. Note that no intra-cavity hard aperture is used. The soft aperture, resulting from the spatial gain modulation, is the dominating mode-locking mechanism.

The dependence of the pulse duration on the position of the prism (P2) was experimentally verified and the results are depicted in Figure 2.4. Three different regimes can be distinguished (A, B, and C). The first regime (A) encloses the pulses ranging from 35 fs down to 20 fs. A linear behaviour between the prism position (linear proportional to the negative group delay dispersion) and the pulse duration is observed, as expected from Eq.(2.4). For the 30-fs pulse, the spectrum, non-collinear autocorrelation and modeshape are given in Figure 2.4b\((a)\). The laser cavity mode has a near-Gaussian profile as can be inferred from the CCD image. The laser spectrum is situated on the infrared side of the spectral range. With a reduction in the GDD the spectrum shifts towards the visible side, and broadens but its shape remains Gaussian-like. Further decrease of the overall net group delay dispersion in the cavity results in the appearance of a bi-stable (hysteresis) behaviour in the pulse generation. (Figure 2.4 marked B and C). Following the trajectory B, the pulse duration first
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Fig. 2.4. (a) Pulse duration as a function of the relative prism position (P2). Autocorrelation traces were measured using a non-collinear arrangement employing a thin (30µm) BBO crystal. The pulse duration is obtained from a fit to the autocorrelation trace (sech²(t) pulse shape). Trajectories A, B, and C denote the hysteresis curve which is observed when the prism position is tuned. The dashed vertical line marks the position of zero group velocity dispersion (D=0). The position where the shortest pulses (optimum conditions) can be obtained is marked by the large grey circle. Note that the appearance of pulses with pulse duration shorter than 10 fs is an artifact and is ascribed to the fit to the center of the autocorrelation function, thereby fully ignoring the wing structure in the trace. (See for example the data marked by γ).

For three settings of the prims position (marked by α, β, and γ), the laser-mode-shape, non-collinear autocorrelation and laser spectra are given (b). Mode profiles were measured by a CCD camera, and the logarithm of the intensity is depicted. The spectra were recorded by an optical multichannel analyzer (Princeton Instruments Inc.).
Cavity-dumped Titanium:Sapphire laser

increases followed by a sudden jump to a low value. The downward jump is accompanied by a laser cavity mode change. With the increase in the pulse duration, the autocorrelation acquires long wings, and is not described by a sech\(^2\)(t) pulse shape any more. For the point marked $\beta$, the mode, pulse, and spectral characteristics are depicted in Figure 2.4b($\beta$). The laser mode now exhibits a more diffusive profile, and a small low intensity satellite ring can be observed.

With the downward jump several changes in the laser characteristics are observed. The diameter of the central part of the laser mode shape is slightly reduced. Furthermore, the spectrum of the laser exhibits a double peaked structure, and is accompanied by a significant pulse degradation, as can be inferred from the appearance of strong side wings in the autocorrelation function. A fit to the central part of the correlation function thereby ignoring the side lobes, yields values as short as 7.5 fs. For the most extreme case ($\gamma$), the laser and pulse parameters are depicted in Figure 2.4b($\gamma$).

Upon the introduction of more prism material in the laser cavity (more GDD), the side wings in the autocorrelation function disappear and the pulses gradually broaden up to 35 fs. The trajectory $C$ is in this case followed. With the increase of the net negative GDD, the double peaks in the spectrum slowly merge, resulting in a bell-shape spectrum. The optimum pulse conditions resulting in the shortest pulses without significant wing structure are generated near the first bending point of the curve $C$ (prism positioned at -0.25 mm).

Operation of the laser closer to the zero group velocity boundary results in a significant pulse distortion (Figure 2.4b($\gamma$)). The setting of the laser to the optimum shortest (and cleanest) pulse condition is thus accomplished by first over-compensating the laser (positive GDD) and then slightly introducing a net negative GDD up to the point that the output spectrum of the laser exhibits a broad structure, peaking at 780-790 nm with a longer tail to the infrared side. The remarkable appearance of the bi-stability of the laser near the zero GDD boundary is attributed the presence of higher (third, fourth) order contributions in the overall GDD of the cavity.[38,61,62,75-77] The double peaked spectrum and associated pulse distortion have been reported in several papers, and seem to be a characteristic of the prism controlled Ti:sapphire lasers.[38]

The influence of the third order dispersion has been calculated by Brabec et al.[38,59] and from the theoretical simulations, a similar behaviour is found as measured in Figure 2.4a, trajectory $C$. The pulse duration drops faster than the linear behaviour, as a result of the presence of the third-order dispersion. Most probably the strong Kerr-nonlinearity in combination with the complicated wavelength dependence of the group delay dispersion might explain the multi-stability of this cavity.[78] The operation of the laser above the zero GDD point (overcompensation) results in the generation of long picosecond pulsed radiation.[79]

Figure 2.5 shows the interferometric autocorrelation and the spectrum of the shortest pulses obtained from the laser. The autocorrelation was measured by conventional collinear
Fig. 2.5. Interferometric autocorrelation (a) and spectrum (b) of the femtosecond laser pulses from the cw-mode-locked Ti:sapphire laser. The dashed curves in panel (a) depict the resulting envelopes of a calculated fit to autocorrelation of a sech²(t) pulse shape. The resulting fit value for the pulse duration is Δτ = 10.5 fs. The inset shows the layout of the real-time autocorrelator setup used to record the interferometric trace depicted in panel (a). The incoming beam is split in two, one beam is directed through a variable delay, whereas the other beam has a fixed delay. Both beams are recombined on the second beamsplitter and focussed with a lens in a thin BBO crystal. The second harmonic radiation is directed to the photodiode detector, after removing of the fundamental radiation by a filter. The autocorrelation scanning repetition rate is 10 Hz.

The second harmonic generation in a 30-µm thick BBO crystal, employing a Mach-Zehnder interferometer configuration (Fig. 2.5, inset). The use of identical thin beamsplitters inside the interferometer, ensures that both pulses, as they arrive at the non-linear crystal, have experienced an identical optical path and thus the same group velocity dispersion. As can be inferred from Figure 2.5a, clean pulses, free of any substantial wings were obtained. The autocorrelation function was fitted to the theoretical expected shape (a pulse with a sech²(t) intensity profile) and resulted in a pulse duration of 10.5 fs. The dashed lines represent the calculated envelope for the interferometric fringes. The measured laser output spectrum is depicted in Figure 2.5b. The spectrum is characterized by a full width at half maximum (FWHM) of 75 nm. The appearance of a shoulder on the IR (long wavelength) side of the spectrum is a result of the coexisting multiple regions of negative group velocity dispersion separated by region with zero or even positive GDD, and is caused by the presence of higher order terms in the group velocity dispersion function. The measured value of the time-bandwidth product is equal to ΔτΔν = 0.4, and slightly larger than the expected value for a sech²(t) pulse (0.315). The deviation is a result of the non-sech shape, which leads in general to a larger value of the time-bandwidth product (Gauss: ΔτΔν = 0.441). The absence of substantial wings in the interferometric autocorrelation function is a good indication that the residual chirp in the pulse is negligible.
After the initiation of the mode-locking process, the laser persisted operating in the short pulse mode. Interruption of the mode-locking occurred whenever the intra-cavity radiation was blocked (for instance, by a large dust particle) or whenever the laser experienced a strong external perturbation (acoustic shock). Note, that the last effect can also be used to initiate the short pulse generation process. Enclosing the laser cavity with a cover, thereby reducing the air (dust) flow through the laser cavity, strongly improved the stability characteristics of the laser.

2.4 Cavity-dumped mode-locked Ti:sapphire laser

Although the pulse characteristics of the cw-mode-locked Ti:sapphire laser are well suited for numerous types of optical studies, several serious drawbacks are encountered in the direct application toward non-linear spectroscopic experiments. The major restriction is the high repetition rate (~80 MHz) and the high average power of the output pulse train. In general, optical experiments should be performed in a single shot limit and with a low average power (thermal load) applied to the sample. Furthermore, non-linear optical studies would greatly benefit from a larger energy in the pulse. Amplitude of the laser pulses at lower repetition rates is a commonly followed route to reduce the repetition rate and increase the pulse energy. However, this would require the implementation of a complex amplification chain.

A considerable simpler approach is the process of cavity-dumping of the laser. In this process, the energy stored in a high-Q cavity is suddenly released. The switching of the optical radiation out of the cavity can be performed by either acousto-optical (Bragg refraction) or electro-optical (Pockels effect) methods. In principle both methods can be applied to cavity-dump a femtosecond Ti:sapphire laser. However, from the viewpoint of the additional dispersion introduced in the laser by the optical switch, the acousto-optical method is preferred, because of the short optical length and good dispersion properties of the Bragg cell.

The successful incorporation of an acousto-optic cavity-dumper in a Kerr-lens mode-locked Ti:sapphire laser was first reported by Ramaswamy et al. Shortly later we reported on the cavity-dumping of the cw-mode-locked 10-fs Ti:sapphire laser. Following this route, the implementation of a cavity-dumper in a saturable absorber mode-locked Ti:sapphire laser, as well as in a femtosecond mode-locked Cr:Forsterite laser was reported. The application of the electro-optic method of cavity-dumping has recently been shown by Gibson et al.

The layout of the acousto-optic cavity-dumped Ti:sapphire laser is depicted in Figure 2.6. Its configuration is similar to that of the cw-mode-locked laser (Fig.2.3), be it that the short (non-dispersive) arm of the laser is now equipped with a Bragg cell, placed in a secondary
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fold. The so-called double pass energy extraction mechanism is applied, which results in a better extraction efficiency from the cavity. Specific details on this interferometric dumping scheme are presented in Appendix 2A. In order to compensate for the additional dispersion of the Bragg cell, the intra-cavity prism separation of 54 cm in the 4-mirror version of the laser is lengthened to 73 cm in the cavity-dumped configuration.

A pick-off mirror separates the two parallel beams (cavity mode and cavity-dumped beam) and directs the cavity-dumped beam towards a four-prism dispersion compensator, for (pre)compensation of additional elements in the setup further on.

Let us now consider the new stability conditions of this 6-mirror laser cavity. With the addition of a second fold, the one dimensional dependence as given in Figure 2.2b, changes into a two dimensional plot, where the stability ranges now depend on the distance between the Ti:sapphire crystal focusing mirrors (M1, M2) as well as the distance between the cavity-dumper mirrors (M3, M4). Figure 2.7 shows the stability diagram of the 6-mirror laser configuration, which was calculated using the ABCD-matrix formalism. Two regions can be distinguished where stable laser operation is possible. Note that similar stability characteristics have been found for the 6-mirror linear-hybrid mode-locked femtosecond dye lasers.

Similar as in the calculations presented in Figure 2.2, the action caused by the presence

Fig. 2.6. Schematic of experimental set-up for cavity-dumping of a Ti:sapphire laser. Ar* - argon-ion laser (Coherent Innova 310); L1 - f=12.5 cm lens; Ti:S - 4 mm Ti:sapphire crystal (Union Carbide); Bragg cell - 3 mm fused-silica acousto-optic modulator (Spectra-Physics, Model 344, Bragg cell); HR - high reflector (CVI); M1-M2 - R=10 cm mirrors (Spectra-Physics); M3-M4 - R=10 cm mirrors (LEZER-OPTIKA); OC - T=3% output coupler (LEZER-OPTIKA); M5 - pump beam steering mirror; Pick-off mirror, M6 - gold coated mirrors; P1-P6 - fused silica prisms; RF driver - cavity-dumper driver (Spectra-Physics, Model 454); PS - Periscope for beams steering and polarization rotation. Note that although the cavity-dumped beam is depicted as being displaced in the plane of the optical table, in reality it travels above and below the cavity mode.
Fig. 2.7. Stability diagram of the cavity-dumped Ti:sapphire laser. The stability zones I and II are within the shaded areas. The distances of the mirror separations of the most stable Kerr-lens mode-locking configuration are indicated by the solid black area along the inner border of the second stability zone. The dashed lines depict the stability ranges of the resonator in case a Kerr-lens (f=30 mm) is assumed inside the Ti:sapphire crystal. The distances between the fixed cavity elements are: (HR-M4)=205 mm; (M3-M2)=355 mm; (M1-P1-P2-OC)=1060 mm.

of the laser field on the material parameters is modelled by simulation of a lens inside the Ti:sapphire crystal. Upon the insertion of this Kerr-lens, a significant change in the stability zones is found. Especially the inner border of the second (II) stability zone experiences a large shift. Operation on the inner border of the second stability zone should give a preferential gain for the high intensity mode-locked mode. Experimental verification showed that the most stable mode-locking occurred along the inner border of the zone II (thick solid line in Fig. 2.7). Furthermore, as has been established earlier, this configuration has a lower resonator sensitivity to external perturbations. The distance between the mirrors M3 and M4 was chosen to be near confocal (dot-dashed horizontal line in Fig. 2.7) in order to maintain the original position of the mirrors M1 and M2.

In cavity-dumped dye lasers the output coupler is conventionally replaced by a high reflector, thereby minimizing the cavity losses. The increased cavity Q-factor results in a larger circulating intra-cavity power. However, the closing of the cavity in the Ti:sapphire laser led to the effect of multiple pulse formation. The generation of two (or even more) highly synchronized short optical pulses, is then sustained. A small output coupling efficiency can prevent this effect to occur. The highest cavity-dumped output and shortest pulses were obtained with the T=3% output coupler.

For triggering of the cavity-dumper electronics, the auxiliary beam passing through the output coupler is directed to a fast photodiode. The pulsed Ti:sapphire laser itself acts as the master oscillator and all further electronics are slaved to its mode-locking frequency. Specific details on the RF driver electronics are presented in Appendix 2B.
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Fig. 2.8. Interferometric autocorrelation (a) and spectrum (b) of the cavity-dumped pulses. The pulse duration is 12.8 fs assuming a \( \text{sech}^2(t) \) intensity profile. The dashed lines in panel (a), represent the calculated envelope for the interferometric fringes.

The self-mode-locking process was started by moving prism P2 back and forth in the cavity or by tapping the output coupler (OC, Fig. 2.6). The RF modulation provided by the cavity-dumper was not sufficient to start the laser at a stable basis. When the laser had optimum alignment, the pulses from the side of the output coupler were less than 13 fs long and had an average power of 200 mW at a 82-MHz repetition rate, when pumped with 5.5 W. From this we can conclude that the additional material and optical elements added to the original design just slightly change the performances of the original cw-mode-locked laser, with a small increase in pulse duration from 11 fs to 13 fs.

Figure 2.8 shows the interferometric autocorrelation of 62-nJ dumped pulses at a repetition rate of 80 kHz along with their spectrum. The interferometric traces were measured using an autocorrelation setup as described before (Fig. 2.5 inset). A fit assuming a \( \text{sech}^2(t) \) shape for the intensity envelope yields a pulse width of 12.8 fs and provides a good representation of the experimental data. The measured time-bandwidth product of 0.38 is reasonably close to the theoretically predicted value of 0.315 for a \( \text{sech}^2(t) \)-shaped pulse. The absence of any significant sidebands in the autocorrelation signal in Figure 2.8 shows that the discrepancy between the measured and calculated time-bandwidth product is not due to residual chirp on the pulse but rather to a pulse shape that deviates slightly from a \( \text{sech}^2(t) \) pulse shape. As is usual the case for prism-controlled Ti:sapphire lasers,\(^{37,38}\) the spectrum exhibits some asymmetry with a tail towards longer wavelengths. Further decrease of the amount of negative group velocity dispersion in the cavity, by moving one of the prisms, led to rapid degradation of the pulse quality.\(^{39}\) The dynamics of the laser pulse durations on the magnitude of the net negative group delay dispersion (GDD) for the cavity-dumped
configuration shows a similar behaviour as depicted in Figure 2.4. The spectra and autocorrelation functions of the pulses coming either through the output coupler or via the cavity-dumper were identical within experimental uncertainty. Inspection of the spectrum across the diffracted beam showed that no transverse frequency chirp occurred. This accords with the fact that the divergence of the beam tightly focussed into the acoustic-optic cell exceeds, even for 10-fs pulses, the variation in the angle of diffraction caused by the bandwidth.

Figure 2.9a depicts the signal from a fast photodiode monitoring the dumped pulse train. The measured contrast ratio between the dumped and preceding/trailing pulses is at least 150:1,[88] while the pulse-to-pulse stability was estimated to be about 1% rms. The buildup dynamics of the pulse energy inside the resonator at a repetition rate of 80 kHz, as was monitored on the side of the output coupler, is depicted in Figure 2.9b. The estimated (double pass) dumping efficiency as obtained from the ratio between the cavity depletion and the steady state level was approximately 80%. This value is consistent with an estimate of 75% as derived from the ratio between average powers and repetition rates of the dumped and intra-cavity pulses. Higher dumping efficiencies resulted in the failure of the synchronization electronics to trigger the RF driver properly.

The transient dynamics depicted in Figure 2.9b capture some intriguing aspects of the pulse generation in a Kerr-lens mode-locked laser. Due to the significant reduction of the intra-cavity pulse power after the dumping event, the saturation of the gain is relaxed. With the pump rate being constant an exponential initial growth of the intra-cavity energy should be expected. However, the intra-cavity pulse energy remains almost constant for a period as long as 50 round trips. This fact means that self-amplitude modulation, caused by the gain-spatial profile due to longitudinal pumping, plays an essential role in the laser. A decrease
in the intra-cavity pulse energy reduces the self-focusing of the cavity mode thereby increasing the diameter of beam waist in the gain medium. As a consequence the spatial overlap between the cavity mode and pump beam decreases leading to a drop in the gain. It is only after some time, because of accumulation of population inversion in the gain medium, the pulse energy starts to grow considerably, going through an overshoot\cite{56,57,83} before relaxing to the steady-state level.

The modelling of the transient dynamics of the cavity-dumping process is presented in Appendix 2C. The simulations take into consideration both the linear and nonlinear losses, as well as the population dynamics. The numerical results are found to be in good agreement with the observed dynamics.

At repetition rates higher than 200 kHz the laser could also be stably mode-locked but with lower dumping efficiency. For instance, at 400 kHz the pulse energy was typically 50 nJ, but at 4 MHz it has decreased to 20 nJ. In case the high dumping efficiencies are maintained at high repetition rates the laser ceased mode-locking. Figure 2.10 presents data on the average power and the energy in the pulse for different repetition rates. Inspection of the transient behaviour after the dumping event and the relaxation back to the steady state conditions (Fig. 2.11b), a cavity recovery time of 4 µs can be deduced. From this, the maximum dumping frequency for the largest efficiency is estimated to be in the range of 250 kHz. For higher dumping repetition rates, smaller dumping efficiencies must be applied. This to ensure that the intracavity laser power returns to the steady state level before the next dumping event takes place. Note that for all repetition rates the measured pulse duration deduced from the interferometric autocorrelation (analogous to Fig. 2.8a) remains 13±1 fs.

Fig. 2.10. Pulse energy (solid circles) and average power (solid triangles) versus dumping repetition rate. Results are given for dumped pulses with duration of 13 fs.
2.5 Tunability of the cavity-dumped laser

In the previous section we have utilized the full accessible bandwidth of the laser for the generation of pulses with the shortest duration. With these ultrashort pulses many optical experiments can be performed. However, several classes of experiments rely on a finite bandwidth of the excitation pulses. For instance, in dynamical holeburning experiments the bandwidth of the laser must be smaller than the width of the absorption band probed.[90] Wavelength dependent pump-probe experiments, as commonly used in the study of relaxation processes in biological functional units, profit from the tunability of the laser source.[91] Different parts of the broad absorption band can be probed this way. Furthermore the ‘narrowband’ operation of the laser allows the selective wavepacket excitation for example in semiconductor quantum well structures.[92]

In this paragraph it is demonstrated that with a minor modification, the bandwidth of the laser can be restricted, allowing the generation of longer and tunable laser pulses. To do so, a variable-width slit is inserted at the dispersive end of the laser cavity near the output coupler (Fig. 2.6). The width of the slit determines the spectral bandwidth, whereas the position of the slit sets the centre wavelength of the laser emission. The effect of reduced bandwidth operation inherently results in lengthening of the pulse.

Although the inclusion of the tunability option can be installed in both the 4-mirror laser cavity as well as the 6-mirror cavity-dumped version of the laser, the latter configuration is only discussed. In the long pulse regime, the dumped pulses exhibit some residual chirp. External compensation with fused-silica prisms resulted in impractical prism separation distances. Therefore, the fused silica prisms were replaced by prism made of a slightly more dispersive material (LaFN28). After passing this prism arrangement, nearly transform limited pulses are obtained. The reason of the presence of the residual chirp on the cavity-dumped pulses is yet unknown.

Fig. 2.11. Tunability of the cavity-dumped mode-locked Ti:sapphire laser. In this experiment, the pulse duration was deliberately set at 27±3 fs over the whole tuning range. The cavity-dumping repetition rate is 200 kHz. The dashed lines serve as a guide to the eye.
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With the insertion of the slit in the cavity, (tunable) pulses with durations ranging from the full bandwidth limit (13 fs) up to a duration of 45 fs can be generated. A typical example of such a long pulse mode of operation is depicted in Figure 2.11, where the pulse duration is deliberately kept around 27±3 fs. The tunability of the centre wavelength of the laser can be changed from 750 nm to 860 nm, and is restricted by the wavelength-dependent dielectric reflection properties of the mirrors used in the cavity. Note, that for different choices of the centre wavelength, different slit positions as well as different slit-widths must be applied, to generate pulses with a 27-fs pulse duration. The corresponding laser spectra have Gaussian shapes. Near the limits of the tuning range a small asymmetry in the spectrum is observed. As a result of the changing GVD versus wavelength, each new position of the slit requires a slight adjustment of the intra-cavity as well as extra-cavity dispersion compensation prisms.

In conclusion, the long pulse option of the laser forms a valuable extension to the short pulse operation of the laser, and turns the short pulse laser in a versatile tunable tool for spectroscopic purposes. Switching between the short pulse and long tunable pulse mode of operation, is rapidly accomplished by the simple insertion of a variable-width slit in the cavity.

2.6 Non-linear optical experiments

By cavity-dumping the 10-fs Ti:sapphire laser, a light source is now available that can directly be applied to numerous non-linear optical studies. In this section we will address some applications of the radiation from the cavity-dumped Ti:sapphire laser.

First, the applicability of the laser toward the problem of solvation dynamics in the liquids is presented. Two different classes of four-wave mixing spectroscopy will be discussed, pump-probe and photon echo. Making use of the cavity-dumped pulses, it is shown that the four-wave mixing signals from dissolved dye molecules can easily be generated. Although the specific dynamical features of the measured transients are extensively analysed and discussed in the following chapters, the aim of this section is to demonstrate the advantages of the use of a cavity-dumped laser over the cw-mode-locked laser. The issue of repetition rate dependence and power dependence will be addressed.

In the second part of this section, the white-light continuum generation in a single mode fibre is discussed. This broadband continuum can be recompressed to pulses with sub-10-fs duration. Furthermore, we demonstrate that the broadband continuum can be used as excitation pulses in a chirped four wave mixing experiment.
2.6.1 Pump-probe spectroscopy

In a pump-probe experiment, changes in the transmission induced by the interaction with a relative strong pump pulse are probed by a weaker probe pulse. The pump-probe experiment is primarily sensitive to the population dynamics, although the presence of any vibrational coherence might appear as quantum beats in the signal.

In Figure 2.12 the pump-probe signals for both cw-mode-locked excitation and the cavity-dumped pulse train are depicted. The transient as measured for the low repetition excitation rate, is typical for a pump-probe experiment. No signal is observed at negative delays, and an induced transmission can be seen after the pump pulse has interacted with the sample. The result obtained with the cw-mode-locked pulse train shows a significant distortion from the cavity-dumped case. One might argue that the signals for delays longer than $t_{12}=100$ fs are identical, apart from a scaling factor, but around zero delay, as well as for negative delays, a large discrepancy is observed between the two signals measured at different repetition rates. The occurrence of these anomalies is a result of the non-single shot character of the experiment performed with the cw-mode-locked laser. The possible presence of thermal gratings and accumulation of population in long-living triplet states are avoided in a low repetition rate experiment where every pump-probe combination excites a fresh sample spot. The inter-pulse separation in case of the cw-mode-locked experiment is in the order of 12 ns, and judging from the results depicted in Figure 2.12 this time is not sufficient to have the system recovered back to its initial configuration. Furthermore, the accumulation of photochemical product states is avoided in a reduced repetition rate experiment.

![Fig. 2.12. Comparison of a pump-probe study performed with cw-mode-locked laser pulses (80 MHz, dashed line) and cavity-dumped operation (400 kHz, solid line). The sample is a carbocyanine dye HITCl dissolved in ethylene glycol, and applied in a fast flowing jet. The inset shows the layout of the experimental arrangement. Typical pulse energies in this experiment are 500 pJ. The pulse duration of the excitation pulses amount 13 fs.](image-url)
2.6.2 Photon echo spectroscopy

Besides the pump-probe experiment, the photon echo is another widely used technique for probing dynamical processes in various systems. \textsuperscript{96-104} In this section, we will show that in the two pulse echo experiment, it is also necessary to use the cavity-dumping laser to obtain reliable data, free of thermal and multiple-pulse effects. Furthermore, the increased pulse energy results in a better signal to noise ratio. In photon echo experiments, the major noise contribution arises from the scattering on irregularities on the jet surface into the direction of the signal. With the noise proportional to the intensity \((N=1)\), and the signal proportional to the cube of the intensity \((S=1)\), the signal to noise ratio scales as \(S/N=I^2\). Increase of the pulse energy (intensity) results in a better signal to noise. However, a large increase in the pulse intensity can result in the appearance of higher order non-linearities. This effect will be illustrated for a series of two-pulse echo signals where it is shown that the enhanced energy per pulse can introduce higher (fifth) order effects, thereby pushing the experiment conditions beyond the third order limits.

In Figure 2.13 the results of a two pulse echo for different repetition rates are given. Similar as in the pump-probe case, the causality in the experiment prohibits the generation of signal before the first pulse (wavevector \(k_1\)) has excited the system. This in general leads to asymmetric profiles as is observed for the cavity-dumped case. Contrary to the low repetition rate experiment, excitation of the sample with the full pulse train results in nearly symmetric echo traces. Already for negative times a signal can be observed. A single shot

![Two Pulse Photon Echo Signals](image)

**Fig. 2.13.** Two pulse photon echo signals measured at a repetition rate of 400 kHz (solid line) and 80 MHz (dashed line). The sample is a carbocyanine dye HITCI dissolved in ethylene glycol. The inset shows the layout of the phase-matching arrangement. The echo-signal is detected in the phase-matched direction \(2k_1-k_2\). Typical pulse energies in this experiment were 500 pJ. The pulse duration of the excitation pulses is 13 fs. The specific details of this experiment are dealt with in Chapter 3.
Fig. 2.14. (a) Two pulse photon echo signals measured for different excitation energies. The energy of the first excitation pulse is changed, the corresponding values for the energy per pulse are indicated along the traces. The intensity of the second pulse is fixed to a value of 0.45 nJ. For clarity the traces are displaced along the vertical axis. (b) Low intensity excitation (1.5 nJ, solid line) and high intensity excitation (9 nJ, dashed line) signals. The intensity of the signals are scaled to match the trailing part of the signal.

limit is thus of a vital importance in echo experiments, to measure reliable data.

Several series of experiments, performed with cw-mode-locked Ti:sapphire laser, have been reported where the full repetition rate of the laser was used for excitation. In most of the data, distortions as described here, have been observed. Reduction of the repetition rate is thus essential for obtaining trustworthy data. For instance, in experiments on bacterial light harvesting complexes (LH2) as well as on photosynthetic reaction centres, is has been demonstrated that the observed data is extremely sensitive to the excitation repetition rate.

Apart from the reduction of the repetition rate, the cavity-dumping of the laser also yields an enhanced energy per pulse. By increasing the pulse energy both the echo signal as well as the signal-to-noise ratio (S/N) grow. However, above certain excitation powers severe changes in the signal shapes are encountered. This is demonstrated in Figure 2.14 where a series of two-pulse echo signals is given for different excitation pulse energies. The full accessible power from the cavity-dumped laser is in many cases too high and induces higher order non-linear processes. Strong distortions of the signals especially around zero delay where the pulses overlap in time, are evident in Figure 2.14.

In the right panel (Fig. 2.14b) a low intensity and high intensity signal are compared. Although the trailing edge of both transients, if scaled properly, can be made to overlap, near the peak position of the echo signal a large discrepancy is seen. Varying the intensity of the other beam resulted in a similar signal behaviour. This shows clearly that, apart from the
reduction of the repetition rate of the laser, intensity dependent measurements of the signal shapes must be performed, to check for the correct power dependence.

Based on the aforementioned findings one may conclude that the low-level pulse energies (<0.5 nJ) are sufficient to generate reliable echo data. For coherent experiments one does not need to cavity-dump the laser. A pulse-picker, installed after the cw-mode-locked laser can easily provide the required pulse energies. However, as is shown in the following chapters, as soon as the coherent techniques are extended towards multiple-pulse experiments, including processes such as signal-gating by means of upconversion techniques, the signal benefits from the high intensity in one of the beams, without thereby distorting the power-sensitive third order signal. In Chapter 3 to 6, the large number of optical elements, causes a significant part of the initial available power to be obsolete. In this case the flexibility in the initial pulse energy as provided by the cavity-dumped laser, is quite useful for a proper performance of the experiment.

2.6.3 White-light continuum generation, pulse compression and chirped four-wave mixing

To demonstrate the potential of the cavity-dumped Ti:sapphire laser for ultrafast spectroscopy, 13-fs pulses of up to 40 nJ at 400 kHz were injected into a 2.75 µm core diameter single-mode polarization-preserving fibre (Newport, F-SPV) of ~3 mm length. The layout of the setup is depicted in Figure 2.15a. The total throughput of the fibre setup was 60%. No damage of the fibre was observed up to the highest pulse energies available. The spectrum of the pulses at the exit of the recollimation lens (L2) extends from 500 nm to 1100 nm (Fig. 2.15b). Comparison to the input laser spectrum, a strong spectral broadening of the initial pulse is observed, which is attributed to the self-phase-modulation in the fibre. The spectral broadening does not occur when the output of the cw-mode-locked laser is injected in the fibre, just a slight change in the spectral width of 10%-20% is observed. The intensity dependence of the spectral bandwidth of the fibre-output exhibits a square-root dependence on the input power, in accordance with the predictions of Tomlinson et al. According to this theory, the output of the fibre should be a linearly chirped pulse, the instantaneous frequency of the pulse changes linear in time. Deviation from the linear behaviour results from higher order dispersive effects in the fibre material and the intensity dependence of the group velocity dispersion (optical shock term).

An attempt was made to shorten the chirped optical pulse to its bandwidth-limited value employing the double grating compressor and 4-prism compressor as was used by the 6-fs experiment of the Shank group. For the compression we used the experimental setup as depicted in Figure 2.15c. The throughput of the complete setup, as a result of the low diffraction efficiency of the grating pair (in total four passes over the grating) was only a few
Fig. 2.15. (a) Schematic layout of the fibre-chirping setup. L1, L2 - microscope objectives. (b) Optical spectra of a dumped pulse before (dashed curve) and after (solid line) passage through 2.7 µm core single-mode fibre. (c) Layout of the pulse compression setup. G1,G2 - gratings; RM - roof mirror, P1-P4 - fused silica prisms. (d) Background free autocorrelation of the compressed pulse (solid line). In this panel, also the fit to pulse with a sech\(^2\)(t) intensity envelope (dashed line) and the Fourier-transform of the spectrum (dash-dotted line) are given.

The total compressor was optimized to obtain the shortest pulse. The non-collinear autocorrelation of the best recompressed pulse was measured and is depicted in Figure 2.15d. Although the (Fourier-limited) minimum pulse duration for this spectrum amounts ~3.5 fs, a fit of the autocorrelation with a sech\(^2\)(t) pulse shape result in a duration of ~7.5 fs. The factor of two difference is a result of the presence of higher-order dispersion contribution which is not properly compressed for. Furthermore, as can be inferred from Figure 2.15d the central part of the autocorrelation function is accompanied by multiple side peaks.

Further developments in the compression of these chirped pulses, thereby carefully characterizing the white-light continuum, and using this information to optimize the compression parameters, have lead to the generation of 5-fs pulses.\cite{117,118} Implementation of chirped mirrors in the compressor, replacing the grating pair has boosted the throughput characteristics of the total compressor without changing the pulse length. Continuum generation and compression techniques with high power amplified pulses has also recently been demonstrated, where a hollow fibre technology was used for generation of the continuum.\cite{119}
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Fig. 2.16. Two pulse echo signal measured with chirped pulse excitation. The sample is the IR-dye molecule HITCI dissolved in ethylene glycol. The inset shows the phase matching arrangement. The tail of the echo response is magnified by a factor of 20. The estimated chirp rate in this experiment amounts $b = 8\ \text{THz/fs}$.

The uncompressed chirped white-light continuum can directly be applied in chirped four wave mixing studies.\textsuperscript{120,121} The experimental arrangement is analogous to the two pulse echo but this time the chirped pulses as they are obtained from the output of the fibre are used as excitation pulses. The sweep in the instantaneous frequency of the pulse relative to the internal timescales of the system dynamics, determines whether the system experiences an impulsive or quasi-cw excitation.\textsuperscript{121}

The echo trace of a chirped four wave mixing experiment is depicted in Figure 2.16. An asymmetric trace where the leading edge is steeper than the falling edge, indicates the presence of slow system dynamics relative to the applied chirp-rate. The appearance of a recurrence in the signal at delay times around 130 fs is a result of coherent Raman scattering processes involving a high frequency vibrational resonance.

Note that this class of experiment cannot be performed with the cw-mode-locked laser. The necessary peak power at the input of the fibre to initiate the spectral broadening process, exceeds the typical values as obtained from a cw-mode-locked system. This class of experiments can either be performed with an amplified system or a cavity-dumped system.

2.7 Concluding remarks

In this chapter a cw-mode-locked prism controlled Ti:sapphire laser generating ultrashort femtosecond laser pulses has been described. Depending on the residual net group delay dispersion in the cavity, the pulse duration can be varied between 10.5 fs and 35 fs. The
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The mode-locking mechanism of the laser is based on the Kerr non-linearity in the Ti:sapphire crystal.

A cavity-dumper that allows the extraction of the intracavity radiation was implemented in the cw-mode-locked laser design. The inclusion of this cavity-dumper changes the operation characteristics of the laser only slightly, bringing the shortest pulse duration to 13 fs. Dumping efficiencies of 80% can be reached at low repetition rates. Even at these high dumping efficiencies, the laser persisted mode-locking. The maximum pulse energy of the dumped pulses can be as much as 60 nJ at a repetition rate of 200 kHz. The cavity-dumped laser of this design was reproduced in many laboratories.[122]

With a slit installed inside the resonator of the cavity-dumped laser, tunability is gained at the expense of pulse duration. A tuning range over 100 nm is possible for pulses with duration of 27±3 fs.

The main virtue of the cavity-dumped laser over the cw-mode-locked version is the reduced repetition rate, allowing single shot experiments. Furthermore, higher intensities can be utilized for purposes of signal upconversion, and multiple pulse experiments. For several non-linear four wave mixing experiments, (pump-probe and photon echo) it is demonstrated that cavity-dumping of the laser is essential for measuring signals free of distortions, that arise as a result of the accumulation of heat, triplet-state population or photo-products.

The multi-kHz and sub-MHz repetition rates as they are used throughout this thesis ensure an efficient data-collection and averaging. Moreover, implementation of signal processing methods based on lock-in amplifier and phase sensitive detection can be used. This in combination with the excellent stability (intensity and pulse duration) of the femtosecond laser source yields for many experiments good signal to noise ratios. The robustness and durability of the cavity-dumped Ti:sapphire laser makes it a reliable laser source for non-linear optical studies.

Repetition rate and power dependent experiments as presented in Figures 2.12, 2.13, and 2.14 help to select the correct probe to be used in solvation dynamics studies. Different probes show different sensitivity toward the appearance of higher order distortions in the signals. Furthermore, the decay rates of the transients that serve as a possible probe of the solute-solvent interactions differ from probe to probe. From comparative studies under several carbocyanine dyes it follows that the dye DTTCI encloses excellent characteristics, such as the relative long decay, a large insensitivity to multiple pulse effects as well as high excitation energies.

Upon injection of the high intensity short pulses in a fibre a white-light continuum can be generated that can be directly used for non-linear optical spectroscopies. This chirped continuum can be further recompressed to sub-10-fs pulses by known pulse compression techniques. The white-light continuum in combination with current achievement in the pulse compression techniques, enhances the time-resolution in time-domain spectroscopy. The combination of the ultrashort pulse duration and the large spectral width further broadens the

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range of possible applications. Making use of spectral filtering techniques, the output of the fibre can be modified into several tunable pulses.[123] Multiple wavelengths are available that extend over a significant tuning range. Furthermore, the coherent nature of the continuum makes it an attractive near-white-light source, with intrinsic fibre coupling, to be used as excitation source in optical interferometric tomography.[124,125] The axial resolution in this technique is determined by the coherence length of the excitation light which for this source amounts ~2 µm. Also in the field of the two-photon excitation confocal microscopy, this laser has drawn some considerable interest. The enhancement in the axial resolution and the strong reduction in the background signal show great potential toward a better resolution.[126-129]
Appendix 2A.

Double-pass cavity-dumping

The schematic layout of the cavity-dumper (acousto-optic Bragg) placed inside the laser resonator is depicted in Figure 2A.1.\(^{[130]}\) As the pulse that circulates in the cavity enters the Bragg cell, part of the radiation is scattered on the RF-induced refractive index grating under a specific angle.\(^{[130],[131]}\) Note that the layout of the cavity-dumping configuration is analogous to that of the Michelson interferometer, where the beamsplitter can be regarded to be replaced by the Bragg cell.

For the description of the dumping process the optical pulse is assumed to be given by:

\[
E(t) = E_0 \cos(\omega t) \tag{2A.1}
\]

where \(E_0\) denoted the time dependent envelop and \(\omega\) the laser carrier frequency. The scattering of the light on the RF-induced sound wave causes a frequency shift of the pulse by an amount equal to the RF frequency \((\Omega)\). The expressions for the scattered pulse \(E_s\) and the depleted cavity pulse \(E_d\) after the first pass through the Bragg cell are given by:

\[
\begin{align*}
E_s(t) &= \sqrt{\eta_1} E_0 \cos(\omega t + \Omega t + \Phi) \\
E_d(t) &= \sqrt{1 - \eta_1} E_0 \cos(\omega t)
\end{align*} \tag{2A.2}
\]

where the intensity diffraction efficiency is denoted by \(\eta_1\), and \(\Phi\) is the adjustable phase of the RF carrier.

---

Fig. 2A.1. Schematic layout of the double-pass cavity-dumping arrangement. For the definitions of the electric fields see text.
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After the pulse has traversed the Bragg cell for the second time, the total extracted pulse is given by the sum of the two scattering contributions as:

\[ E_{\text{tot}}(t) = \sqrt{1 - \eta} \sqrt{\eta} E_0 \left[ \cos(\omega t + \Omega t + \Phi) + \cos(\omega t - \Omega t - \Phi) \right] \]  

(2A.3)

Similar as in the Michelson interferometer, the output depends on the specific phase relation between the two adding pulses. The resulting intensity of the dumped output of the laser is:

\[ I_{\text{damped}} = 4 \eta (1 - \eta) |E_0|^2 \cos^2(\Omega t + \Phi) \]  

(2A.4)

The dumping efficiency can be enhanced in case the interference is constructive, or decreased for destructive interference. Note that the full efficiency of 100% can be reached, for a single pass dumping efficiency of 50% (\( \eta = 0.5 \)) and proper setting of the phase \( \Phi \), (\( \Phi = 0 \)). This situation is identical to the Michelson interferometer where the intensity can be fully switched from zero to maximum for a T=50% beamsplitter and proper optimized phase.

As a result of the fact that the envelope of the RF pulse (~rise/fall time of 7 ns) has more or less the same duration as the cavity round trip time (12 ns), the suppression of the preceding and trailing pulse presents a crucial issue in the dumping process.\(^{112}\) Efficient suppression of the pulse can be performed by locking the RF carrier frequency to the repetition rate of the cavity-dumper. From equation (2A.4) it can be inferred that for a phase setting of \( \Phi = 0 \), and a RF frequency \( \Omega \) equal to \( \Omega = (n + 1/2) \nu_c \), the preceding and trailing pulses are fully suppressed, where \( \nu_c \) denotes the laser repetition frequency.

In conclusion, the double-pass configuration allows an efficient extraction of the pulsed cavity radiation even with relative low powers applied to the Bragg crystal. Furthermore, when the phase of the RF frequency is locked to the laser mode-lock frequency, efficient suppression of both the trailing and preceding pulses is obtained.
Appendix 2B.

Cavity-dumper electronics

As was presented in Appendix 2A, the RF frequency (and phase) of the cavity-dumper driver should be slaved to the laser oscillation frequency. Furthermore, the exact timing of the dumping event should be synchronized to the laser pulse train. For the triggering of the RF cavity-dumper driver, we use the auxiliary beam passing through the output coupler (T=3%, Fig. 2.6), and direct it to a fast photodiode. Since the action of cavity-dumping causes the intra-cavity power to drop significantly (Fig. 2.9b), a chain of two amplifiers is used, to ensure a significant signal level for triggering of the cavity-dumped electronics at all times. A schematic layout of the synchronization electronics is given in Figure 2B.1. For the matching of the repetition rate of the laser (~82 MHz) and the input of the cavity-dumper driver circuitry (~41 MHz) an additional divider by two is installed. The cavity-dumper driver unit (Spectral-Physics Model 454) allows the setting of the timing of the RF pulse, as well as the adjustment of the phase of the RF carrier with respect to the RF pulse envelope. The latter is used to phase-synchronize the dumped pulses in the double extraction scheme, as was explained in Appendix 2A. The bandwidth of the internal phase-locked-loop (PLL) of the cavity-dumper driver unit, imposes a limited range in the acceptable repetition rates, and thus in the length of the cavity-dumped laser. In the design employed here, the round trip length was set to 1.83 m, resulting in a repetition rate of 82 MHz.

Fig 2B.1. Simplified block diagram of the synchronization electronics. Laser - cavity-dumped Ti:sapphire laser; Diode - fast photodiode (silicon); Amp1, Amp2 - 200-MHz, 20-dB amplifier; f/2 - 'divide by 2' digital circuit; RF - Cavity-dumper driver electronics (Spectra-Physics Model 454); Bragg cell - fused silica Bragg cell (Spectra-Physics Model 344S cavity-dumper).
Appendix 2C.

Transient dynamics in the cavity-dumped Ti:sapphire laser

In this appendix the transient dynamics of the cavity-dumped Kerr-lens mode-locked Ti:sapphire is addressed. The simulation of the transient features, are based on the following laser rate equations, which can be derived for the photon number \( n \) and the population inversion \( N \):\(^{4,5}\)

\[
\frac{d n}{d t} = K(n+1)N - \left[ \gamma_e + \gamma_{KLM}(n) \right] n
\]

\[
\frac{d N}{d t} = R_{pump} - K n N - \gamma_{pop} N
\]

In the above equations, \( K \) is a scaling constant and \( \gamma_e \) encloses the resonator coupling losses. The pump rate is represented by \( R_{pump} \) and \( \gamma_{pop} \) denotes the population relaxation time. The Kerr-lens induced losses are included through \( \gamma_{KLM}(n) \). Since the Kerr-lens depends on the intensity of the optical field, \( \gamma_{KLM}(n) \) has a photon number dependence. The exact form of the intensity dependence strongly depends on geometrical factors (cavity configuration and pump spatial conditions). In the analysis presented here, we will use an S-shape functional behaviour that fulfills the following boundary conditions. High photon numbers, corresponding to a large intensity, should lead to a decrease in the KLM losses, whereas low intensity should give large KLM losses, as was discussed in Section 2.2.2.

In Figure 2.C1 the results of the numerical simulations are depicted for three different dumping efficiencies (\( \eta = 20\%, 50\%, \text{ and } 80\% \)). Two special cases for the Kerr-lens losses are discussed. Besides a system where the Kerr-lens induced losses are taken into full consideration, a second simulation is performed for a system where the latter effect is excluded (\( \gamma_{KLM}(n)=0 \)).

For the low dumping efficiency, the influence of the Kerr-lens induces non-linearity in the laser recovery dynamics is nearly zero. The results for both simulations are only slightly different. After the 20\% of the cavity photons are dumped, a gradual, nearly exponential growth is seen that gradually reach the steady state level.

Inspection of the traces for dumping efficiencies as big as 80\% shows that the significance of the presence of the Kerr-lens induced phenomena is striking. Inspection of the bottom panel shows that the Kerr-lens effect is as big as the output coupling losses. Upon the dumping of the cavity power, the low photon number causes the Kerr-lens induced losses to increase immediately. The response time of this non-resonant
Fig. 2.C1. Computer simulations of the relaxation dynamics after cavity-dumping of the Kerr-lens mode-locked Ti:sapphire laser. The upper three panels display the photon number dynamics for three different dumping efficiencies, $\eta = 20\%$, $\eta = 50\%$ and $\eta = 80\%$. The middle row contains the population inversion number $n$ and the lowest row depicts the Kerr-lens action (losses) $\gamma_{\text{klm}}$ relative to the output coupling $\gamma_{\text{out}}$. The photon number dependence of the Kerr non-linear damping rate $\gamma_{\text{klm}}$ is depicted in the inset of the lower left panel. The suffix ss denotes the steady state value.

The mechanism compared to the cavity trip round trip time can be regarded as infinitely fast. With the increase of the losses, in combination with the low photon number, the depletion of the inversion, as a result of the process of stimulated emission, is only small. With the pump rate being constant, the inversion grows steadily up to the point that the overall gain surpasses the total losses. The presence of a far above steady state inversion in the system causes eventually a strong exponential growth in the photon number. Even an overshoot in the photon number above the steady state level, can be observed.\textsuperscript{[56,37,44]} Simultaneously with the growth in the photon number, the Kerr-lens losses decrease. After some time the system reaches back to the
steady state. Depending on the pump level, strong relaxation oscillation can be observed in this case. Here the parameters are chosen in such a way that a damped behaviour is observed.

When comparing the $\eta=80\%$ photon number result with the presence of the Kerr lens and without the presence of the Kerr-lens, a significant change in the transient behaviour is seen. A direct growth in the non-Kerr case versus a plateau or even decaying photon number in the presence of the Kerr-losses. Inspection of Figure 2.9b shows that the intra-cavity pulse energy remains almost constant for a period as long as 50 round trips. This fact means that the Kerr-lens self-focussing plays an essential role in the laser. Note that for the large dumping efficiencies (80%), a longer relaxation time (3µs) is needed, for the laser system to reach the steady state level.

The relaxation oscillation as observed in Figure 2.9b, are not observed in the calculations shown, but have also been observed in theoretical numerical simulation on the pulses propagation in the Ti:sapphire laser. The latter simulations consider the space-time focussing of the short optical pulse into full detail.\[65\] Oscillation both in the pulse width as in the spectral bandwidth are associated with the relaxation oscillations. This fact might result in a situation that at certain time after the dumping event, a pulse with a duration shorter than the steady state condition is propagating in the resonator.

Based on the above model, the calculations clearly capture the observed transient dynamics of the cavity-dumped laser. Evidently the Kerr-lens induced losses form a crucial factor in the relaxation dynamics of the cavity-dumped laser after each dumping event.
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66. In the linear method as presented by Ref. 35, the operators are considered to be commutative. In the (3D,t) calculations the order of the non-commutative operators are treated as they are encountered in a round trip through the cavity.


72. The author gratefully thanks Prof. H.C. Kapteyn and Prof. M.M. Murnane from the Washington State University for providing us details on the cw-mode-locked Ti:sapphire laser and sending us a copy of Ref. 71.

73. It is a great pleasure to acknowledge F. Krausz (Technische Universität Wien, Abteilung für Quantenelektro und Laser) for his invaluable comments on the laser design and operation.


78. The bistable behaviour has also been observed by I. Shumay, Universität Erlangen, Nürnberg, Germany, private communication (1996).


80. A similar Mach-Zehnder interferometer design is used in the preparation of phase-stabilized pulses. For a detailed description see Chapter 4, Section 4.4.

81. We are indebted to Prof. J.G. Fujimoto for providing us with the preprint of Ref.56, and for helpful comments on the installation of the Spectra Physics cavity-dumper.


88. Note that the previously reported (Ref. 57) value of 50:1 was obtained with an older version of the Spectra Physics driver electronics. After the upgrade of the driver electronics the contrast ratio has increased to 150:1.

89. The maximum single pass efficiency of the cavity dumper is ~40%.


A cavity dumped laser based on the presented design are currently in use or have been used in the following laboratories. N.F. Scherer, University of Pennsylvania, Philadelphia, Pennsylvania; G.R. Fleming, University of Chicago, Chicago, Illinois; M. Joffre, ENSTA, Ecole Polytechnique, Palaiseau, France; G.J. Brakenhoff, University of Amsterdam, Amsterdam, The Netherlands; T. Aartsma, Leiden State University, Leiden, The Netherlands; A.L. Gaeta, Cornell University, Ithaca, New York; I. Shumay Univeritat Erlangen-Nurnberg, Erlangen, Germany; J.G. Fujimoto, Massachusetts Institute of Technology, Cambridge, Massachusetts; K. Yosihara, Institute for Molecular Science, Myodaiji, Okazaki, Japan.


Spectra Physics, preliminary instruction manual, Model 344S cavity-dumper & Model 454 cavity-dumper driver.


Note that nowadays cavity-dumper drivers can be purchased, that have standard build-in constant-fraction discriminator and divider/2 electronics. (Camac, Model CD4000) Furthermore, this unit has a short RF pulse (~9 ns) that is applied to the crystal, ensuring a better contrast ratio for the preceding and trailing pulses. (1000:1, private communication, M.S. Pshenichnikov, B. Hesp 1997).

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